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ZIRCONIUM CHLORIDE

NUCLEAR BATTERY

by

Lonnie J. Shalton

A

Thesis

Submitted to the Faculty of the

University of Missouri School of Mines and Metallurgy

in partial fulfillment of the requirements for the

Degree of

Master of Science in Metallurgical Engineering

Rolla, Missouri

1964

Approved by:

Advisor

ABSTRACT

An investigation of the neutron absorption characteristics of zirconium chloride was made to verify the reports of Chicago Development Corporation (5810 47th Ave., Riverdale, Md.) that ZrCl might be used as a power source for a nuclear battery. The company detected what they believed to be a beta current when the ZrCl was bombarded with neutrons from a slightly enriched uranium source. Investigation using higher powered neutron sources revealed that such a current was highly improbable and the company incorrectly interpeted the phenomena. Reports in the literature also confirmed that an electric current from the ZrCl using a low powered neutron source was not likely.

ACKNOWLEDGEMENTS

The author wishes to express his sincere appreciation to Dr. W. J. Kroll for the granting of a fellowship without which this graduate study would not have been possible.

The author is indebted to Dr. D. S. Eppelsheimer, Dr. M. E. Straumanis, Dr. W. H. Webb, Dr. D. R. Edwards, and the nuclear reactor staff for their suggestions and valuable guidance throughout this investigation.

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CHAPTER I

INTRODUCTION

Statement of the Problem.

The purpose of this investigation was to expand preliminary experiments carried on by Chicago Development Corporation concerning the use of zirconium chloride as an atomic battery power source.

In personal correspondence (Jan. 22, 1959), Dr. D. S. Eppelsheimer, Chairman of the Department of Metallurgy at the Missouri School of Mines, received information from Dr. R. S. Dean, President of Chicago Development Corporation, concerning unique electron-neutron absorption characteristics of zirconium monochloride. Dr. Dean stated that it might have use as a power source of a nuclear battery. A sample of the zirconium chloride was sent to Dr. Eppelsheimer by the company with the suggestion that research work might be done at MSM to expand the preliminary experiments.

The zirconium chloride carries the tradename "Zirklor" and is the monochloride form (ZrCl). It is produced by fused salt electrolysis which leaves it relatively free of hafnium, a high neutron absorber.

The nuclear experiment described by Dr. Dean included the following steps:

1. Pressing ZrCl to form cylinder.
2. Hole drilled through axis of cylinder.
3. Ends covered with commercial silver paste for electrical contacts.

4. Electrical circuit set up, containing ammeter to detect current.
5. Bias placed across cylinder to get current reading.
6. Uranium, slightly enriched, inserted into cylinder hole, resulting in increase of current.
7. Battery shorted out, current persisting.

The hypothesis is that the thermal neutrons from the uranium source pass readily through the zirconium layers and are absorbed by the chlorine. The chloride forms, at least in part, isotopes with short half life which disintegrate mostly by emitting beta rays. These high speed electrons are then absorbed in the zirconium layers of the mass; and when a small potential bias is supplied, the compound develops a negative resistance. That is, it becomes an electron source in series with the original bias.

It was reported that the effect was hardly measurable with the weak neutron source used, but with a strong source (e.g. the MSM reactor), the phenomenon might "develop into something".

Organization of the Problem.

The problem was attacked experimentally by preparing pressed ZrCl compacts and attaching electrical contacts for collection of electrons. The compact was then arranged in a clamp to assure complete contact and this apparatus set up in an electrical circuit suitable for placing the ZrCl next to the neutron source. An ammeter was placed in the circuit to detect current changes due to electron emission.

Importance of the Study.

The energy resources of the world are rapidly diminishing and methods of using nuclear power are of growing importance. Nuclear batteries of various types have been prepared, most of them employing a thermocouple arrangement with a semiconductor or using the radiant heat of disintegration in conjunction with a thermoelectric generator to produce energy. This study investigates the possibility of a ZrCl battery involving the direct conversion of nuclear to electric energy, with no intermediate step of heat conversion.

CHAPTER II

LITERATURE SURVEY

Nuclear batteries, as most nuclear energy devices, have been only recently investigated to any great extent. Although many researchers have explored nuclear batteries, there is still no particular type that is generally accepted as the most efficient and economical. Consequently, research is continuing on many such types of devices and materials. The primary supporter for the present research in this area is the U.S. Government, particularly those branches that are involved in nuclear power.

There are four general types of nuclear batteries widely mentioned in the literature:

(1) Directly charged (primary source, alpha or beta emitter) with collector electrode.

(2) Semiconductor (p-n) junction.

(3) Thermoelectric, employing thermocouples absorbing heat produced as radioactivity.

(4) Contact potential, employing isotope to ionize gas in electric field.

The directly charged type of nuclear battery contains an emitter of beta particles which are collected to form a beta current. The region between the emitter and collector is evacuated, although it has been found[1] (1953) that the vacuum may be replaced by a plastic, provided the beta particles are not unduly absorbed. The directly charged battery was first demonstrated by Mosley[2] in 1913 and is probably the earliest proposed nuclear battery.

G. I. Ruckman[3] et. al. (1953) employed a direct cell with a capacitor to accumulate the charge of beta radiation. The S^{35} sulfur isotope with a half-life of 87.1 days was used as a source of beta radiation.

Another direct method of converting nuclear to electrical energy was shown by Guenther[4] (1962). In this case, a core with a fissionable material was in direct contact with an emitter electrode which was electrically insulated from the collector electrode.

In semiconductor (p-n) junction batteries, radiation produces an electron avalanche which is highly efficient in producing current. Rappaport[5] (1954) demonstrated this effect by irradiating germanium and silicon p-n junctions with beta particles from a Sr^{90}-Y^{90} source. Lomer[6] (1954) developed a theory for the emf produced by the semiconductor junction under irradiation.

Pfann and Roosbroek[7] (1954) made an electric power source by exposing the p-n junction to radiation so the junction field separates electron-hole pairs produced by radiation. An 8% efficiency is reported from a Sr^{90}-Y^{90} source.

The electron-voltaic effects produced by doping Ge with As and Ir was investigated in Japan by Yamanaka[8] et. al. (1958). The system was irradiated with a 50 mc Sr^{90}-Y^{90} source and exhibited results similar to solar batteries.

A junction type battery for powering transistor circuits was patented by Rappaport[9] (1963) and was reported to have good efficiency and long life.

Most of the researchers[5,6,8] of junction type batteries report

the detrimental behavior of structure failure of the semiconductor as due to irradiation. Practical use of this type battery necessitates a compromise between long lifetime and high efficiency[5]. Linder [10] et. al. reported that work is underway aimed at alleviating radiation damage (1955).

One of the most recent applications of nuclear power for batteries involves a double conversion: nuclear energy to heat to electrical energy. The thermoelectric type was first shown by Jordan and Birden[11] (1954) and consisted of a Po^{210} source to supply the heat of radioactive disintegration in conjunction with a thermopile to convert the heat to electrical current. The model contained a 150 c source of Po^{210} which then cost $375,000.

K. C. Jordan's work for the Atomic Energy Commission on the thermoelectric battery was also reported in two patents. The earliest[12] (1958) includes two insulating discs with conductive rods dispersed between them to form a circular cage. In the center of the cage is sealed the radioactive source and the thermopile then connected. The other[13] (1959) battery is contained in a capsule which acts both as a shield and a heat convertor. The radioactive decay is converted to useful heat at the capsule surface and this heat is then conducted to hot thermojunctions to give constant current flow to the cold junction.

A thermoelectric generator is used by the Martin Company[14] (1960) for the government's SNAP III program (Systems for Nuclear Auxiliary Power). Many experiments are still being carried on using this setup involving a Po^{210} (and also a Sr^{90}) source as decay heat and the thermoelectric generator as a conversion device.

In the contact-potential-difference cell, the volume between two dissimilar metals is uniformly irradiated by a radioisotope and the resulting ionization is collected by the contact potential between the plates. The phenomenonwas first demonstrated by J. V. Kramer in 1925[15] and was rediscovered for practical use in this country by Ohmart[16] (1951) and others.

Extensive research has been performed on the CPD-type by Tracerlab[17] (1955) under the sponsorship of the U.S. Signal Corps. The primary work has involved gaseous tritium in heavy gases like argon and krypton between various electrode surfaces.

Many other batteries have been investigated and among them are the following:

In Russia[18] (1959), a radiation galvanic cell, based on the oxidation-reduction reaction of Fe^{2+}/Fe^{3+}, is being developed to convert radiation into electrical energy.

An interesting triode concept is demonstrated by Anno and Fawcett[19] (1962) wherein fissionable material on a cathode emits positive charged fragments of high energy and electrons of low energy. The positive ions travel to the anode while the negative grid bias turns the electrons toward the cathode and an electric current is therefore set up.

The possibility of a double conversion with Si photocells where the energy of beta particles is transformed into light and then into electrical energy has been studied[20] (1959). The same phenomenon was involved using a Sr^{90} activated light source[21] (1962) and a photovoltaic nuclear-photon device[22] (1963).

CHAPTER III

THEORETICAL CONSIDERATIONS

Introduction

The proposed zirconium chloride battery source differs from most of the batteries previously investigated in at least one of two respects: the ZrCl battery will not involve an intermediate conversion of energy, and it will not depend on radioactive elements for radiation. The latter factor means that the ZrCl will have to be irradiated by an outside source to form isotopes and then placed into service.

This type of battery is dependent on the half-life of the irradiated material. A very long half-life would require a proportionately long time in the reactor for the material to become energized, while too short a half-life material, although able to be energized quickly, would not give sufficient battery life. The factors to be considered therefore are the beta producing powers of Zr and Cl and the corresponding half-lives.

The theory proposed by Chicago Development Corporation was that thermal neutrons passed through the zirconium layers of the compound and reacted with chloride atoms to form beta rays. These high speed electrons were then absorbed in the Zr layers of the mass and when a small potential bias was supplied, the compound developed a negative resistence and thus became an electron source in series with the original bias. If enough of these electrons could be produced and successfully collected to form a current, an electrical source could develop. If the ZrCl has proper half-life

values, it could be removed from the thermal neutron source and still produce electricity for a length of time.

Reactions

When chlorine is bombarded by thermal neutrons, the primary reaction is

$$Cl^{35} + n^{1} \longrightarrow Cl^{36}.$$

This isotope accounts for 75.53% of the chlorine isotopes formed and decays as a beta producer, but the half-life is 3×10^{5} years. It would therefore take a considerable length of time to charge a battery on the basis of this reaction.

The other reaction involving chlorine,

$$Cl^{37} + n^{1} \longrightarrow Cl^{38},$$

accounts for the remainder (24.47%) of the chlorine isotopes formed and has a half-life value of 37.3 minutes, a possibility for battery use. The decay mechanism of this isotope is

$$Cl^{38} \xrightarrow{37.3 \text{ min}} Ar^{38} + \text{beta particle}.$$

This reaction is therefore the essential electron producer of the chlorine atoms.

The zirconium has a very low cross section and is very ineffective for producing beta rays and hence the governing factor for the electron production from ZrCl is the Cl^{38} isotope of chlorine.

The short half-life of the important isotope obviously limits the ZrCl battery to specific applications. In an area where a nuclear reactor was available only a portion of the time, the ZrCl could be energized periodically without using the reactor for

extended time periods. Energizing a long half-life material would necessitate extended reactor time.

A related application could possibly be where a neutron source must perform intermittently several functions in one operation, as in a space ship. The ZrCl could be charged up to a desired level and then the neutron source could be applied to other operations until the battery needed recharging.

Preliminary theoretical calculations revealed that only a slight current could be obtained from the ZrCl source in the MSM nuclear reactor, but due to the possibility of secondary reactions and the reported results from the Chicago Development Corporation, the investigation was continued.

CHAPTER IV

THE APPARATUS

Zirconium Chloride.

The zirconium monochloride used was obtained from the Chicago Development Corporation (Jan., 1959). From the empirical formula, ZrCl, there is 71% Zr and 29% Cl by weight.

The company prepared the ZrCl by the electrolytic decomposition of $ZrCl_4$ in a fused salt bath. The $ZrCl_4$ contained 1.8% hafnium and 0.2% titanium, but the impurity content for the ZrCl was reported to be

Hf	0.01%
Ti	0.001%
Mn	0.01%
Fe	0.001%
Si	0.001%.

The extraordinary purity of the product is noteworthy, particularly the substantial absence of hafnium and titanium.

Pressing Apparatus.

A die for pressing the zirconium chloride into compact form was fabricated by combining a previously used metal die with a machined steel plug prepared on a lathe. Schematic drawings of the die assembly are shown in Figures 1 and 2.

The body of the die was a cylinder 1 1/2 inches in diameter with a 17/32 inch diameter hole through the center. A base piece fit into the center hole at the bottom of the cylinder to function as the lower pressing face. The pressing plunger was found to be

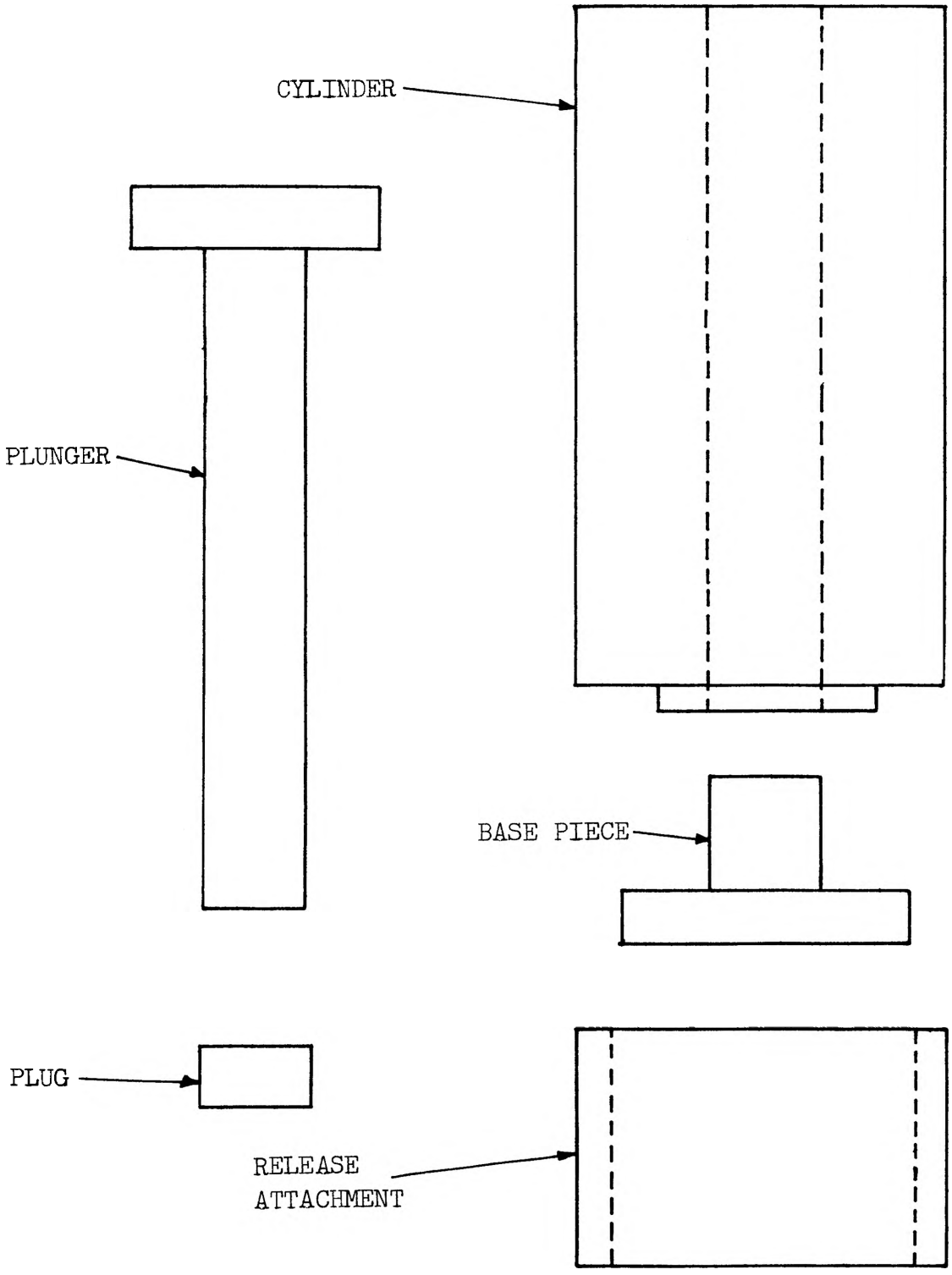

FIGURE 1

PRESSING DIE

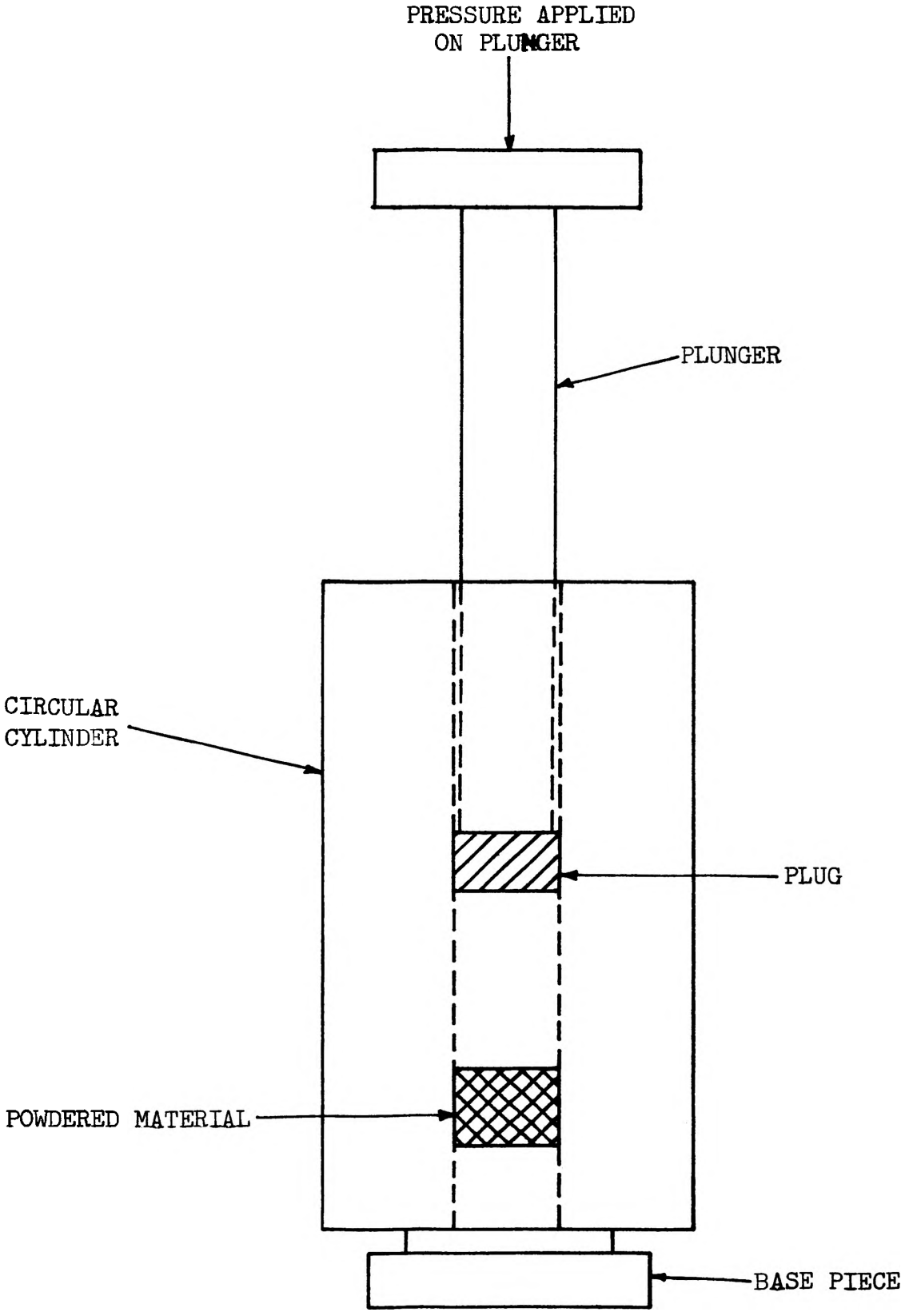

FIGURE 2

DIE ARRANGED FOR PRESSING OPERATION

relatively loose fitting, so a tighter fitting plug was prepared on a lathe. If such a plug was not between the powdered material and the plunger, it was found that the powder would creep up along the side of the plunger under pressure and cause sticking of the die.

A hollow release attachment was provided for removing the compact after pressing.

The procedure for operating the die was as follows:

1. Place base piece in position at bottom of cylinder.
2. Pour material into die and tap die to level off material.
3. Start plug into cylinder.
4. Press plunger into cylinder forcing plug down to material and then press material at desired pressure.
5. Release pressure and place release attachment on bottom of cylinder.
6. Press plunger until base piece, pressed compact, and plug are released at bottom of die.

A simple oil pressure type press was used to make the compacts. The die was placed between two flat plates and the oil pressure was pumped up to a suitable pressure to enable the bottom plate to move up and supply the required pressure to form a compact.

Specimen Clamps.

1. Brass Clamp for Plutonium-Beryllium Source. The specimens for use in this source were arranged in a brass clamp (Fig. 3a) to facilitate handling and to assure good electrical contact.

The specimen and contact materials were arranged in a sandwich fashion, the ZrCl in the center of opposite layers of aluminum foil, aluminum sheet contacts, and plastic insulators. Holes were made in the aluminum sheet extensions (Fig. 3b) for attaching of leads from the electrical circuit. A tight fit was made simply by turning the screw part of the clamp.

2. Wooden Clamp for Nuclear Reactor. The brass clamp used in the Po-Be source was unsatisfactory for use in the nuclear reactor since the brass, upon irradiation by the higher reactor flux, would become very radioactive and potentially dangerous; and a long cooling-off period in the reactor pool would therefore be necessary. An aluminum clamp would have been satisfactory for this purpose, but none was readily available. The need of a clamp presented a dilemma for a short time until a common clothes pin was found to be of virtually a perfect size to handle the specimen.

The final clamp used is shown in Figure 4. The specimen and contact arrangement was the same as in the brass clamp: a sandwich setup with ZrCl in the center, and pressed aluminum foil, aluminum sheet contacts, and plastic pieces on each side of the specimen. The aluminum sheet contained holes in the extension for connecting leads as shown in Figure 3b. In the case of the wooden clamp, the plastic pieces acted as spacers rather than as insulators.

To set up the clamp, the prongs of the clothes pin were held slightly apart and the specimens and contact material placed in

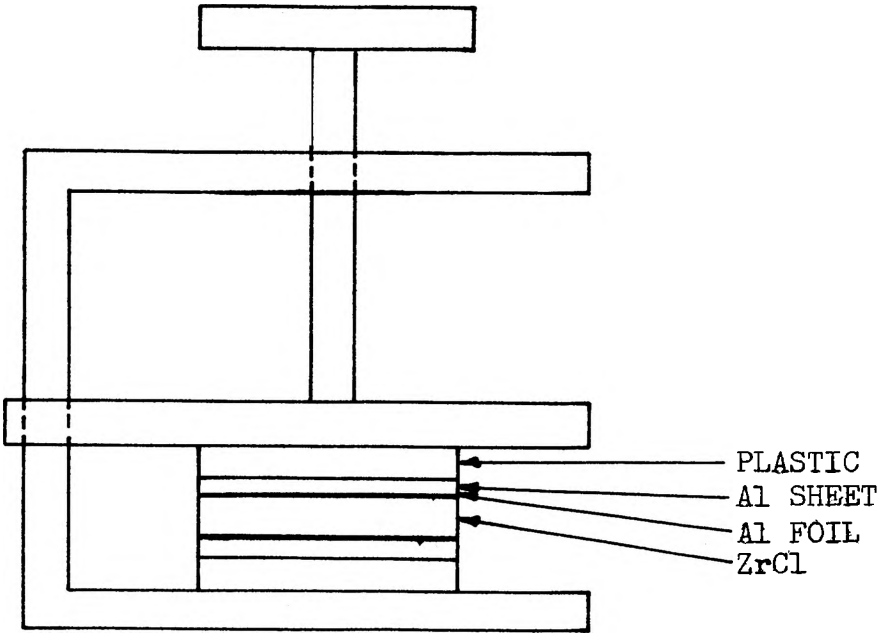

FIGURE 3a

BRASS CLAMP

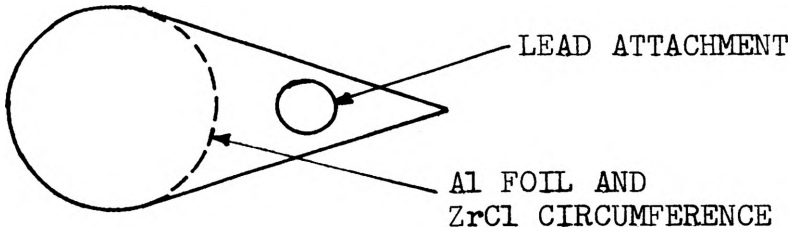

FIGURE 3b

ALUMINUM SHEET EXTENSION

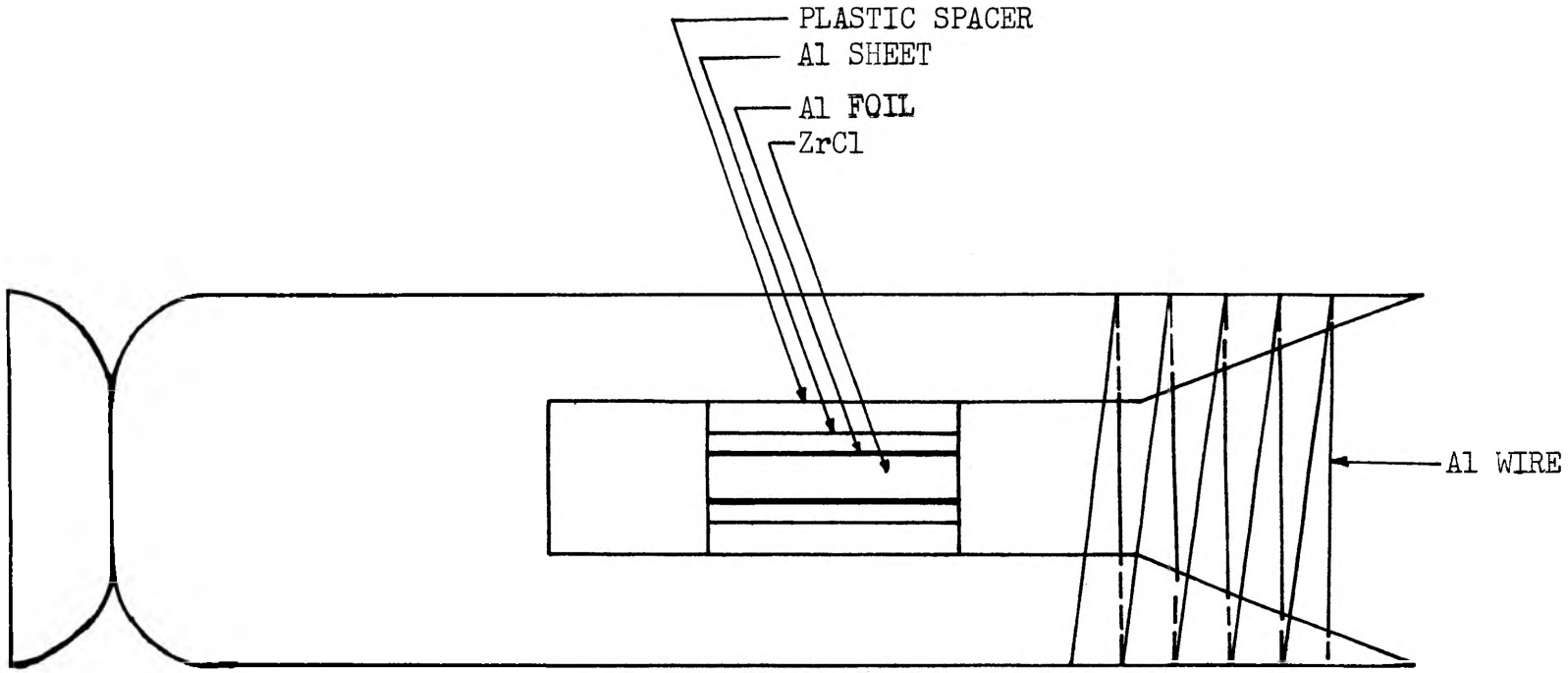

FIGURE 4

WOODEN CLAMP

the center. When the prongs were allowed to spring back, a tight fit resulted. To give more equalized pressure on the specimen, an aluminum wire was wound tightly around the open end of the clothes pin.

Electrical Circuit.

A photograph of the electrical circuit is shown in Figure 5 and a schematic drawing is shown in Figure 6.

The primary parts were a size "D" 1 1/2 volt flashlight battery for bias, an ammeter for detecting current, and a two way switch.

The top terminal of the battery was connected to the two way switch by a wire lead pressing against the top of the battery. A piece of aluminum sheet was shaped to fit at the base of the battery with an extension comimg out to the wire contacts. The battery was held in position by small pieces of wood surrounding it.

When the two way switch at the center contact was placed in position 2, the battery was in the circuit in series with the ammeter and specimen. When the lever was switched to position 1, the battery was by-passed and only the ammeter and specimen were in electrical contact.

The circuit was arranged this way to duplicate the Chicago Development Corporation's experiment with the zirconium chloride. When the specimens were set up with the neutron source, the desired bias could be obtained by switching to position 2. At this position, the ammeter read the combined current produced by

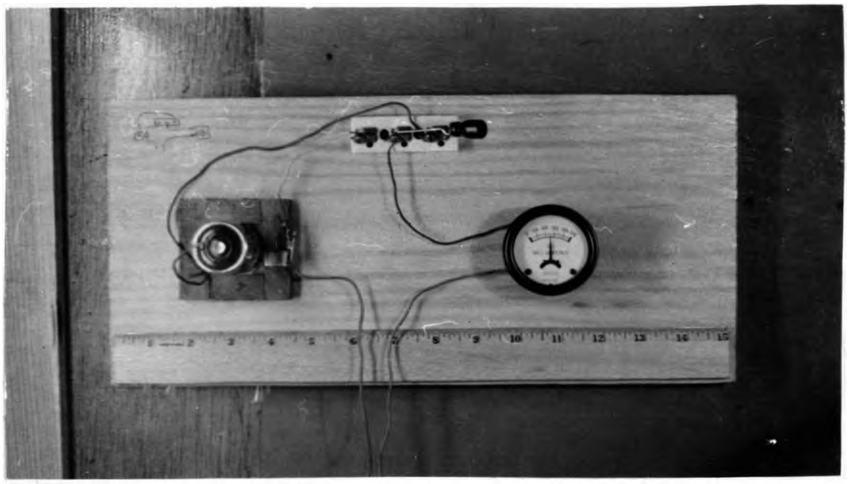

FIGURE 5

ELECTRICAL CIRCUIT

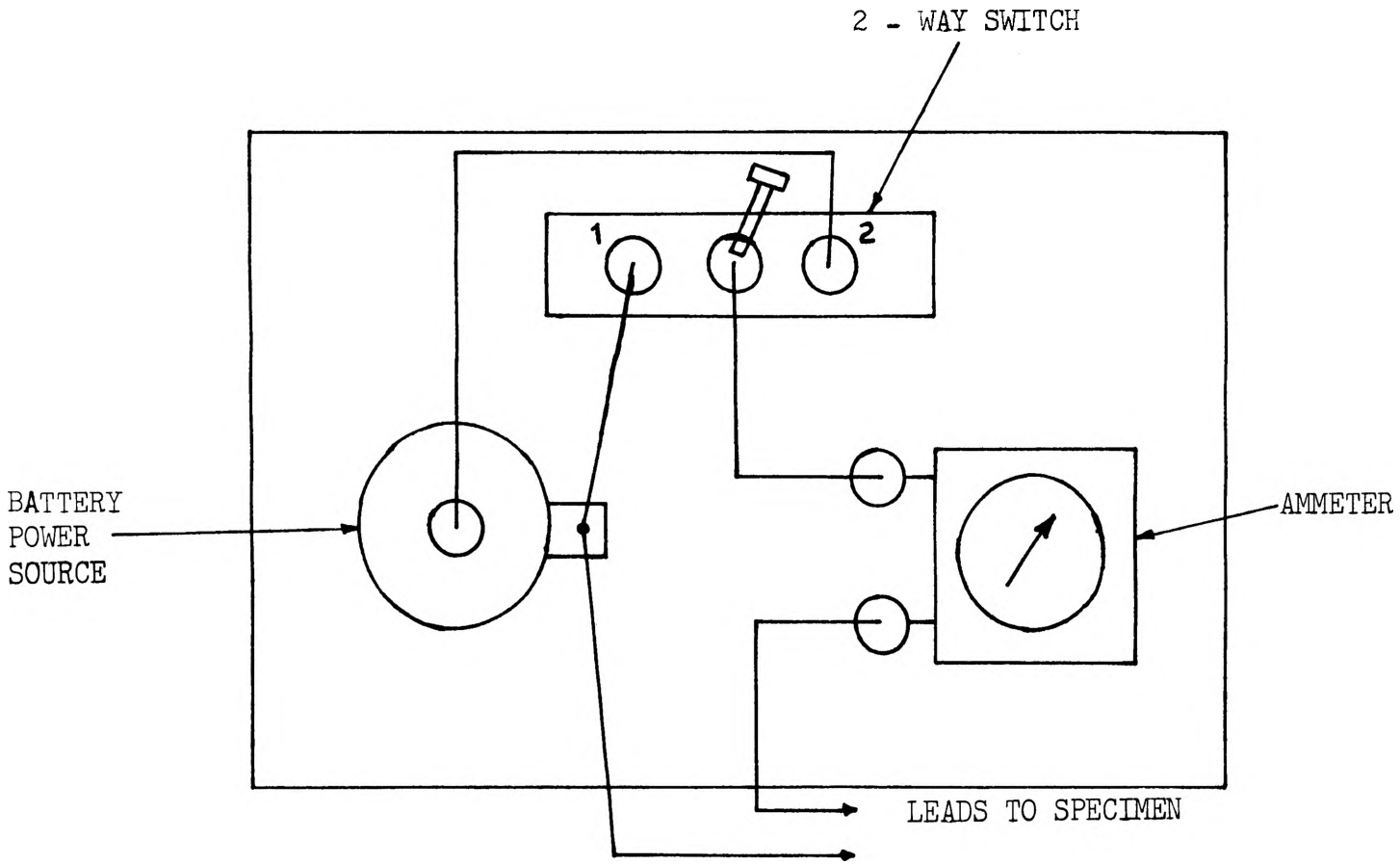

FIGURE 6

ELECTRICAL CIRCUIT

the battery and the electron emission from the ZrCl. When switched to position 1, the only current producer in the circuit was the beta radiation decay of the ZrCl.

Neutron Sources.

1. Plutonium-Beryllium Source. The P -Be source is under the supervision of Dr. W. H. Webb of the Chemistry Department at the Missouri School of Mines. The flux intensity produced is 10^7 neutrons/sec.

The source is immersed in paraffin contained in a large barrel. Two holes are drilled into the paraffin and lead down to the source. Specimens may be placed next to the source by lowering them through the holes.

2. MSM Nuclear Reactor. The Missouri School of Mines nuclear reactor is a heterogeneous, thermal, pool type, research and training reactor. It has a maximum operating level of 10 kw and cooling is by convection.

The pool holds approximately thirty-two thousand gallons of high purity demineralized water. The pool is nine feet wide, nineteen feet long, and twenty-seven feet deep.

The core is composed of fuel elements of uranium-235 plus safety and regulating rods. The maximum neutron flux intensity is 10^{11} neutrons/cm^2/sec.

Specimens were loaded into the reactor by lowering them from a bridge above the pool until the specimen was adjacent to the core. The specimen was attached to a plastic frame which fit into

a notch at the core level.

The reactor is under the direction of a staff headed by Dr. D. R. Edwards.

CHAPTER V

PROCEDURE

Preperation of ZrCl Compacts.

The ZrCl was obtained in flake form, similar to graphite flakes. It was necessary to press the flakes into a compact suitable for handling and attachment of electrical contacts.

Due to the small amount of ZrCl available, the material was divided into two portions: pure, black appearing ZrCl and lighter, oxidized material. To test the pressing characteristics of the ZrCl, the impure material was used in preliminary experiments to avoid waste.

Pressures of 5,000 and 10,000 psi were first tried on samples weighing approximately one gram. The 5,000 psi samples were not compacted sufficiently for handling, but the 10,000 psi samples were suitable for use. A 1.0 gram sample in the 17/32 inch die yielded compacts of 5/16 inch height.

ZrCl specimens throughout the investigation, both pure and impure material, were pressed at 10,000 psi. The weight of 1.0 gram also remained uniform giving compacts of approximately 5/16 inch height.

Electrical Contacts.

A method of successfully collecting the electrons emitted from the ZrCl was needed to detect the reported nuclear phenomena.

Four types of contacts were considered for use: silver paste solution, silver deposited from vapor, pressed aluminum powder, and

thin aluminum foil.

The Chicago Development Corporation described the contacts they used as "commercial silver circuit paste". This type of paste was not readily available however, and other means were sought.

Vapor plating of silver on the compact was considered, but lack of apparatus prohibited this possibility.

Pressing of aluminum powder directly with the ZrCl was tried. A small portion of Al powder was placed in the die and leveled off. The ZrCl was then added, and the plunger was slightly applied to give firmness. Another small amount of Al was then added and leveled off by tapping the die. The specimen was then pressed at 10,000 psi. The product gave poorly attached Al, and the surface was not covered well. Higher pressures and varying amounts of Al were tried but met with relatively little success.

The final method tried was the pressing of common aluminum kitchen foil on the compact. A piece of aluminum foil, shaped to match the die diameter, was placed in the die and forced down to the base piece. ZrCl was added and leveled off. Another aluminum piece was placed on top of the ZrCl and the specimen then pressed at 10,000 psi. A well adhering contact resulted, and the investigation proceeded with this type of contacting arrangement.

Preliminary Electrical Testing.

Preliminary electrical testing was first tried on ZrCl compacts made from the impure material. A series circuit was set up containing an ammeter, power supply, and the ZrCl sample. The compact

of ZrCl was placed on a piece of metal attached to one pole of the power supply while the circuit was completed by touching a wire from the ammeter to the opposite side of the ZrCl.

The first power supply employed was a 12 volt battery in conjunction with a milliammeter. With the circuit completed, no current deflection was obtained.

A 30 volt power supply was next tried, and again there was no current. A microammeter replaced the milliammeter, and a reading of 20 microamperes could be obtained at the maximum 30 volts. The current could be increased to 35 microamperes by squeezing the specimen to get better contact. This led to the use of a clamp as shown in Figure 3.

A compact of pure ZrCl was placed in the circuit, and a current of one ampere was obtained with approximately 10 volts applied.

The currents obtained demonstrated that the electrical contact arrangement was functioning properly and that a bias could be applied in the later experimenting.

It may be noted that when the power source was removed from the circuit, the needle of the ammeter deflected toward the negative direction. This phenomenon may probably be attributed to polarization.

Nuclear Testing.

1. Plutonium - Beryllium Source. All nuclear testing was done with pure ZrCl. For testing with the Pu-Be source, the ZrCl was placed in a clamp (Fig. 3) and then lowered into the source by the electrical leads until it was adjacent to the source. The

leads were placed in the electrical circuit (Fig. 6), bias applied, and current characteristics with and without the bias were tabulated.

2. Nuclear Reactor. Safety requirements set for the reactor necessitated the calculation of activity of the irradiated specimen. Permission for use of the reactor was then granted. The calculations are shown in Appendix 1.

Aluminum wiring, twenty-seven feet long, was used for leads coming from the clothes pin clamp (Fig. 4). After the wires were attached, the clamp apparatus was placed in heated liquid paraffin which was then allowed to cool and harden. The paraffin was applied to assure that electrical contacts would not be interfered with and also to prevent impurities from entering the reactor pool. The clamp was taped to a plastic frame which was then lowered into the pool by the aluminum leads. The frame fit into a notch next to the core which was approximately 25 feet under water. The Al leads were then attached to the electrical circuit (Fig. 6). Two separate experiments were run with the reactor.

A) The first arrangement essentially duplicated the Chicago Development experiment. The paraffin covered clamp was lowered into the pool and placed into position. A bias was applied across the ZrCl with the battery in the circuit and current characteristics due to irradiation were tabulated. The reactor was operated at full power for a time greater than the half-life of Cl^{38}. Reaction for longer than one half-life assured that at least 63% of maximum current would be obtained.

B) A different electrical arrangement was tried on the second

part of the experiment. The two Al sheet extensions in the clamp arrangement (Fig. 4) were both attached to the same lead while the other lead was sharpened to a point and forced into the edge of the ZrCl. Electrons ejecting from the ZrCl would be attracted to the aluminum sheet, leaving the ZrCl positively charged and hence have a bias for electron flow. A battery would therefore not be necessary to establish a current. The completed circuit only contained an ammeter and ZrCl clamp apparatus attached to each other with the aluminum wiring. To screen out secondary electrons that might backscatter and reduce current, a thin piece of plastic sheet was placed between the ZrCl and the Al sheet extensions. The sheet was thin enough so it would not screen out the beta rays. A micromicroammeter was used in this part of the experiment in an effort to detect any current changes. The reactor was operated at full power, and the current characteristics were recorded.

CHAPTER VI

RESULTS

The experimental procedure consisted primarily of three parts: Pu-Be experiment and parts A and B with the reactor.

1. Pu-Be Experiment - With the battery in the circuit as a power source, but before placing the ZrCl adjacent to the source, the current reading was 4 milliamperes on a 100 milliampere maximum deflection ammeter. The ZrCl was placed next to the source for approximately two hours, and the only noticeable change was that the current dropped to 3 milliamperes, probably showing loss in battery power.

2. Reactor, Part A - Due to the length of the aluminum lead wires, the battery gave a current of only 150 microamperes on a 500 microampere ammeter. Using a microammeter enabled the investigator to detect very slight changes in current. The greater sensitivity did not help the results however, as the only change when the ZrCl was placed adjacent to the core was a slight loss in current rather than any gain. The ZrCl was in the reactor for approximately two hours.

3. Reactor, Part B - Since no battery was employed in part B, the only current in the circuit before nuclear activity was due to galvanic action. A micromicroammeter was employed and at the beginning there was a current of 2×10^{-6} amperes. With the reactor at full power for approximately one hour, there was no essential change in current.

To determine if a bias could get something started, the leads

were placed on the poles of a 12 volt battery and then replaced on the ammeter. The current raised to 1.5×10^{-5} amperes but soon receded to near zero current.

As in the previous attempts, the results gave no indication of any nuclear activity.

CHAPTER VII

DISCUSSION AND CONCLUSIONS

The first point to be discussed must be the failure of this investigation to confirm the Chicago Development Corporation's reported results. The problem may be discussed from two viewpoints: 1. The purity of the ZrCl used in the investigation may have deteriorated over the years. 2. The Chicago Development Corporation may have incorrectly interpeted the phenomenon theyobserved.

The first viewpoint is highly possible since the ZrCl near the top of the sample bottle had changed from the pure black color to a white, probably oxidized portion (possibly ZrCl + air $\longrightarrow$ ZrO + Cl_2). The ZrCl used for the nuclear experiments was taken from the black portion of the sample and, by appearance, was pure. The extent of impurity was tested by running specimens of both samples in an X-ray diffraction camera. The diffraction patterns are shown in Figure 7. Although the X-ray films exhibited poor resolution and were not able to be indexed, the strongest line of each pattern was measurable, and the two lines varied sufficiently to indicate two different primary phases. The black material is therefore assumed to be primarily zirconium chloride while the white is probably some form of zirconium oxide.

The other possibility is that of incorrect interpretation. A recent 1964 technical article[23], brought to the attention of the investigator after the above experiments were completed, reported that "A typical rhodium (neutron) detector...produces 0.3 microamperes in a thermal flux of 10^{14} neutrons/cm^2/sec." It appears

highly improbable that Chicago Development Corporation's "slightly enriched uranium", or for that matter the MSM reactor, could have produced a detectable beta current. Since the complete procedure used by Chicago Development is not available, it is very difficult to deduce exactly their observed phenomena.

However, the possibility of a nuclear battery using ZrCl or a similar material cannot be discounted as use with a higher reactor flux may make the operation feasible. Calculations, based on published data[23], indicated that a flux of approximately 10^{17} neutrons/cm^2/sec could produce a current in the microampere range. The calculation is shown in Appendix 2.

In the opinion of this investigator, further research may prove to be valuable.

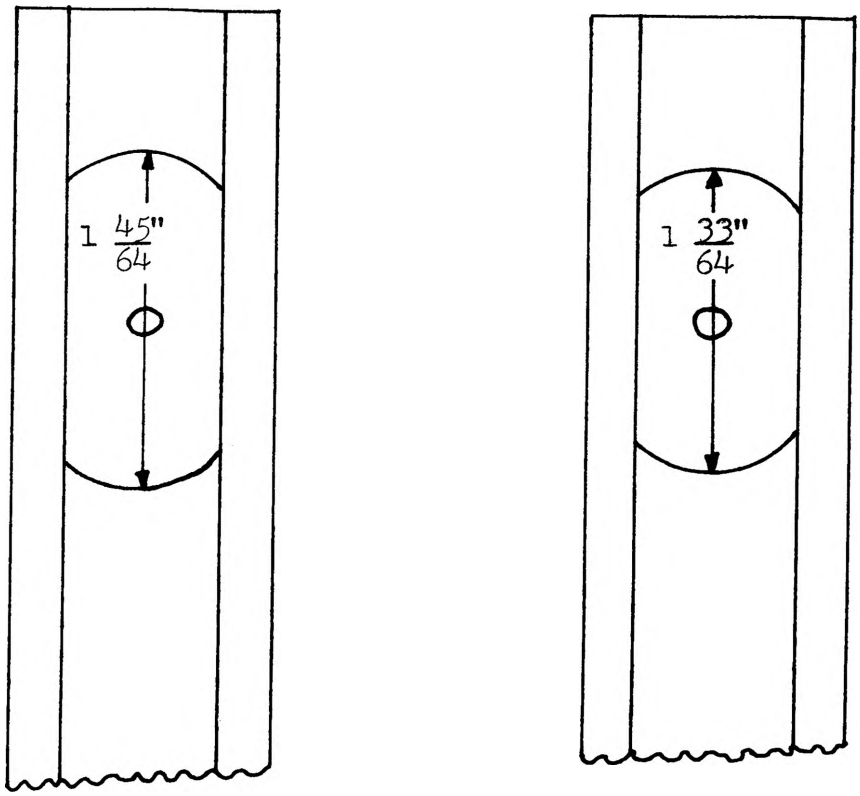

FIGURE 7

DIFFRACTION PATTERNS
(MAXIMUM INTENSITY LINES ONLY)

APPENDIX 1

Activity Calculation of Irradiated ZrCl

DATA

Atomic Weights: Zr - 91.27, Cl - 35.45

Weight of ZrCl: 1.00 gram (0.72 gm Zr, 0.28 gm Cl)

Time in Reactor: 1 hour

Only isotopes emitting gamma rays are considered:

	Cl^{38}	Zr^{93}	Zr^{95}	Zr^{97}
Half Life	37.3 m	20 m	65 d	17 h
σ (barns)	0.56	0.2	0.1	0.1
Per Cent	24.47	17.11	17.40	2.80
E (mev)	4.9	5.0	1.12	2.66

Cl^{38}: N_x = Moles Cl X $\%Cl^{38}$ X atoms Cl/mole

$$N_x = \frac{.280 \text{ g Cl}}{35.45 \text{ g Cl/mole}} \times .2447 \times 6.023 \times 10^{23} \frac{\text{atoms}}{\text{mole}}$$

$$N_x = 1.164 \times 10^{21}$$

$$\lambda = \frac{.693}{T_{1/2}} = \frac{.693}{37.3/60} = 1.115$$

$$\lambda_y N_y = \sigma \phi N_x (1-e^{-t}) = .56 \times 10^{-24} \times 10^{10} \times 1.164 \times 10^{21} (1-e^{-1.115})$$

$$\lambda_y N_y = 4.367 \times 10^{6}$$

$$\text{Curies} = \frac{\lambda_y N_y}{3.7 \times 10^{10}} = \frac{4.367 \times 10^{6}}{3.7 \times 10^{10}} = 1.180 \times 10^{-4}$$

$$\text{Rad/Hour} = 6CE = 6 \times 4.9 \times 1.180 \times 10^{-4} = 3.469 \times 10^{-3}$$

Zr^{93}: $$N_x = \frac{.72 \text{ g Zr}}{91.22 \text{ g/mole}} \times .1711 \times 6.023 \times 10^{23} = 8.134 \times 10^{20}$$

$$\lambda = \frac{.693}{20/60} = 2.079$$

$$\lambda_y N_y = .2 \times 10^{-24} \times 10^{10} \times 8.134 \times 10^{20} (1-e^{-2.079})$$

$$\lambda_y N_y = 1.421 \times 10^{6}$$

$$\text{Curies} = \frac{1.421 \times 10^{6}}{3.7 \times 10^{16}} = 3.8405 \times 10^{-5}$$

$$\text{Rad/hour} = 6 \times 5.0 \times 3.8405 \times 10^{-5} = 1.152 \times 10^{-3}$$

Zr^{95}: $$N_x = \frac{.72 \text{ g Zr}}{91.22 \text{ g/mole}} \times .1740 \times 6.023 \times 10^{23} = 8.272 \times 10^{20}$$

$$\lambda = \frac{.693}{65\text{d} \times 24} = 4.42 \times 10^{-7}$$

$$\lambda_y N_y = .1 \times 10^{-24} \times 10^{10} \times 8.272 \times 10^{20} (1-e^{-4.442 \times 10^{-4}})$$

$$\lambda_y N_y = 3.674 \times 10^{2}$$

$$\text{Curies} = \frac{3.674 \times 10^{2}}{3.7 \times 10^{10}} = 9.930 \times 10^{-9}$$

$$\text{Rad/hour} = 6 \times 1.112 \times 9.930 \times 10^{-9} = 6.673 \times 10^{-8}$$

Zr^{97}: $$N_x = \frac{.72 \text{ g Zr}}{91.22 \text{ g/mole}} \times .028 \times 6.023 \times 10^{23} = 1.331 \times 10^{20}$$

$$\lambda = \frac{.693}{17 \text{ h}} = 4.076 \times 10^{-2}$$

$$\lambda_y N_y = .1 \times 10^{-24} \times 10^{10} \times 1.331 \times 10^{20} (1-e^{-4.076 \times 10^{-2}})$$

$$\lambda_y N_y = 62.557$$

$$\text{Curies} = 62.557/3.7 \times 10^{10} = 1.69 \times 10^{-9}$$

$$\text{Rad/hour} = 6 \times 2.66 \times 1.69 \times 10^{-9} = 2.697 \times 10^{-8}$$

Total Rads/hour = 4.621×10^{-3}

The sample is assumed to be one foot from the source, and the activity is that upon immediate removal from the core. The calculation is based on the reactor operating at the 1 kilowatt level which produces a neutron flux of 10^{10} neutrons/cm^2/sec. At full power, 10 kilowatts, the flux is 10^{11} neutrons/cm^2/sec, and the total activity would by 4.62×10^{-2} Rads/hour.

APPENDIX 2

Flux Intensity Approximation for ZrCl Current Production

Current production due to nuclear reaction is directly proportional to the product of the neutron absorption coefficient, flux intensity, and number of atoms. A ratio may therefore be set up:

$$\frac{\text{Current}_x}{\text{Current}_y} = \frac{\sigma_x \phi_x N_x}{\sigma_y \phi_y N_y}$$

A recent technical article reported[23] a current of 0.3 microamperes using a flux of 10^{14} neutrons/cm^2/sec with rhodium as the activated material. Assuming equal number of atoms and the same current production, the above equation may be used as follows for conversion of Rh calculations to Cl:

$$\frac{\text{Current}_{Rh}}{\text{Current}_{Cl}} = 1 = \frac{\sigma_{Rh} \times 10^{14}}{\sigma_{Cl} \times \phi_{Cl}} = \frac{138 \text{ barns} \times 10^{14}}{.5 \text{ barns} \times \phi_{Cl}}$$

$$\phi_{Cl} = 2.76 \times 10^{16} \text{ neutrons/cm}^2\text{/sec}$$

Therefore, a flux in the range of 10^{17} neutrons/cm^2/sec should produce a current in ZrCl in the microampere range.

BIBLIOGRAPHY

1. Coleman, J. H., Nucleonics 11, No. 12, 42 (1953).
2. Mosley, H. G. J., Proc. Roy. Soc. A88, 471 (1913).
3. Ruckman, G. I., Tychinskiy, V. P. and Yukhvidin, Y. A., "A Method of Producing Power from Beta-Active Isotopes", Trudy Nauch. Isstedovatel. Inst. Ministers tva Radiotekh. Prom. S.S.S.R., No. 6, 3-8, (1956).
4. Guenther, R., Electron Emission Current Nuclear Reactor, Brit. Patent 909,989 (1962).
5. Rappaport, P., Phys. Rev. 93, 246 (1954).
6. Lomer, W., "Direct Conversion of Beta Radiation Energy into Low-Voltage Electrical Energy", A. E. Res. Est., (England) (1954).
7. Pfann, W. G., Roosbroek, W. van, Applied Physics 25, 1422-34 (1954).
8. Yamanaka, C., Wada, H., and Yamamura, Y., Technol. Repts. Osaka Univ. 8, 233-41 (1958).
9. Rappaport, P. (to R.C.A.), Radioactive Batteries, U.S. Patent 3,094,634 (1963).
10. Linder, E. G., Rappaport, P., and Loferski, J. J., International Conference on the Peaceful Uses of Atomic Energy 15, 283-290 (1955).
11. Jordan, K. C. and Birden, J. H., MLM - 984 (1954).
12. Jordan, K. C. (to U.S. AEC), Thermoelectric Generator, U.S. Patent 2,844,639 (1958).
13. Jordan, K. C. and Birden, J. H. (to U.S. AEC), Radioactive Battery, U.S. Patent 2,913,510 (1959).

14. Morse, J. C., "Radioisotopes as Sources of Nuclear Power", Martin Co. TID 7571, 27-38 (1960).

15. Kramer, J. V., The Electrician 93, 497 (1924).

16. Ohmart, P. F., J. Appl. Phys. 22, 1504 (1951).

17. Thomas, A., Quarterly Progress Reports, Aug., 1952 to Oct., 1955, to Power Sources Branch, Components Div., Signal Corps Engineering Lab., Fort Monmouth, N. J.

18. Dolin, P. I. and Duzhenkov, V. I., "Radiation Electrochemical Process", 123-6 (1959).

19. Anno, J. N. and Fawcett, S. L., Battelle Tech. Review 11, No. 10, 39 (1962).

20. Vavilov, V. A., Vul, B. M., Galkin, G. V., and Fridman, S. A., Soviet Physics - Solid State 1, 748-9 (1959).

21. Viczlucky, N., Portable Nuclear Battery, U. S. Patent 3,053,927 (1962).

22. Oestreich, M. C., Robinson, R. J. and Watts, H. V., Contract AF 33(657)-8527 (1963).

23. Hilborn, J. W. Nucleonics 22, No 2, (1964).

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